

Paleo thermal regimes in the  
Yucca Mountain unsaturated zone:

Mineralogical and Fluid Inclusion  
Evidence

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Purpose of the Yucca Mountain secondary  
mineral studies

What is the origin of secondary minerals  
and mineral-forming fluids ?

Rain water percolating through the hot mountain ?

Deep-seated thermal waters injected  
into the vadose zone ?

Subjects

- ❖ Mineralogy and crystal morphology
- ❖ Fluid inclusion temperatures
- ❖ Isotopic properties of calcite
- ❖ A model

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### Secondary minerals found in ESF

**Calcite** -  $\text{CaCO}_3$

**Quartz/chalcedony** -  $\text{SiO}_2$

**Fluorite** -  $\text{CaF}_2$

**Strontianite** -  $\text{SrCO}_3$

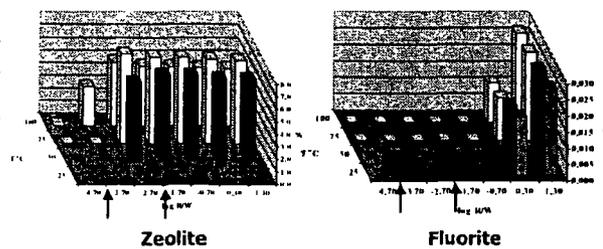
**Apatite** -  $\text{Ca}_5[\text{PO}_4]_3(\text{F}, \text{Cl}, \text{OH})$

**Zeolite (heulandite)** -  $\text{Ca}_4[\text{Al}_6\text{Si}_{28}\text{O}_{72}]24 \text{H}_2\text{O}$

Mineralogy: complex chemistry of fluids

Can complex chemistry of fluids, indicated by minerals be a result of interaction between rain water and tuff ?

### Thermodynamic modeling



Zeolite

Fluorite

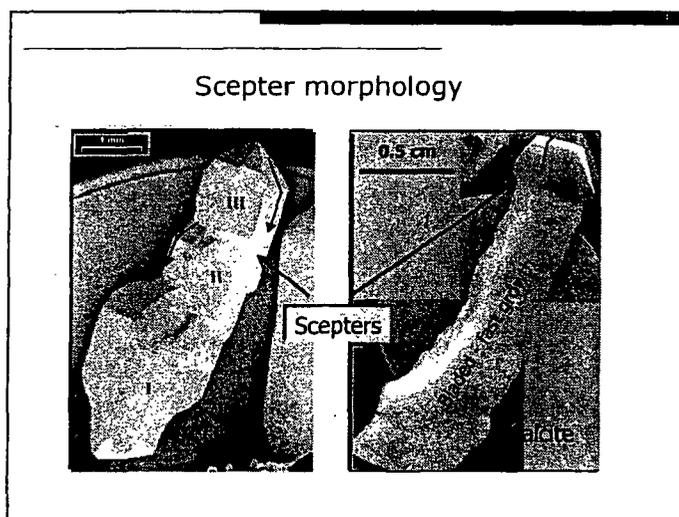
Yucca Mountain rainwater reacts with tuffs;  
Open system ( $O_2$  and  $CO_2$ )

### Mineralogy: Conclusions

- ❑ Complex mineralogy indicates complex and varying in time and space chemistry of water ( $\text{SiO}_2$ ,  $\text{CO}_2$ , F,  $\text{PO}_3$ ,  $\text{SO}_4$ );
- ❑ Minerals that are observed in ESF (e.g., fluorite) should not form from rain water reacting with tuffs; those minerals that should form (e.g., kaolinite, albite, K-feldspar) – are not observed.

### Crystallization in vadose zone from films of water

Can large (cm-scale) euhedral crystals of calcite  
and quartz grow from films of water ?



Crystallization in vadose zone  
from films of water

"... precipitation usually occurs from thin water films that flow over the growing speleothem surfaces. Large crystal terminations do not form on the speleothem surface because they form projections that disturb the water flow away from the projections which, as a consequence, are gradually eliminated."

(Kendall and Broughton, 1978, p. 519)

### Crystal growth from water films: Summary

- No coherent physical model explaining the mechanism of crystallization of large euhedral crystals from films of water has been proposed;
- Examples of growth of large (cm-scale) euhedral crystals of calcite and quartz from films of waters are not known;
- Morphology and growth-related features of crystals from Yucca Mountain indicate growth in submerged state from a fluid with evolving properties.

### Growth rates

"... deposition rates between about 0.035 and 1.8 mm/m.y. are obtained ... These values are in general agreement with long-term rates of mineral deposition during the past 10 m.y. based on direct U-Pb dating of sequential inner layers of opal from calcite-silica fracture and cavity coatings at Yucca Mountain..."

(Neymark et al., 2000)

### Growth rates

In order to ensure growth rate of  
1.8 mm/Ma for a typical crystal from Yucca  
Mountain (e.g., 0.005 x 0.5 x 1.0 cm)  
supersaturation must be maintained at:

$$\Omega = 1.000000045 \text{ to } 1.000000083$$

$$SI = 2.0 \cdot 10^{-8} \text{ to } 3.6 \cdot 10^{-8}$$

### Growth rates

At 50°C:

$$\text{net growth rate} = 2.8 \cdot 10^{-15} \text{ mmol/m}^2\text{h}$$

At 50 + 0.1°C:

$$\text{net growth rate} = 2.8 \cdot 10^{-6} \text{ mmol/m}^2\text{h}$$

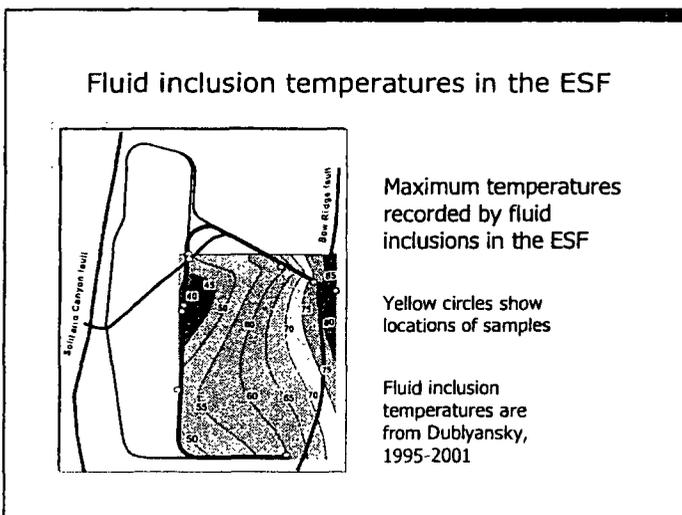
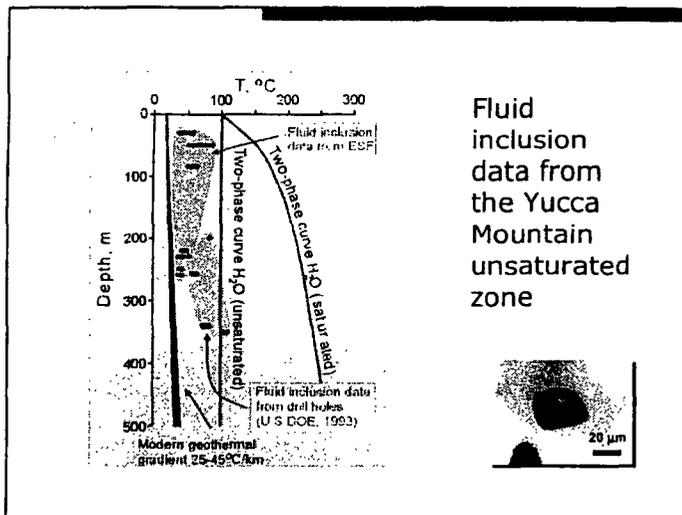
Temperature fluctuation of only 0.1°C  
changes net growth rate by  
9 orders of magnitude.

### Growth rates

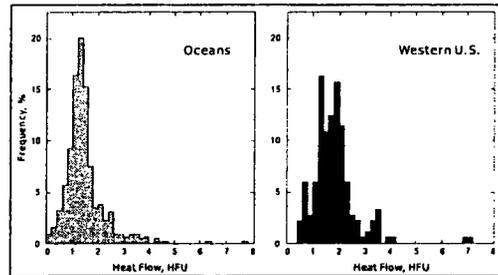
- Deposition rates appear to be unrealistic from the standpoint of general physics, as well as from the standpoint of the theory of crystal growth (inhibition of nucleation).
- This calls into question the results of the radiometric dating.

### Subjects

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- ◇ Fluid inclusion temperatures
- ◇ Isotopic properties of calcite
- ◇ A model

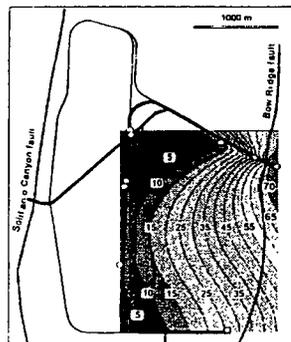


### Heat Flow on the Earth



Heat flow under oceans (by Von Herzen and Lee, 1969)  
and in Western U.S. (by Sass et al. 1971)

### Heat flow based on the fluid inclusion results

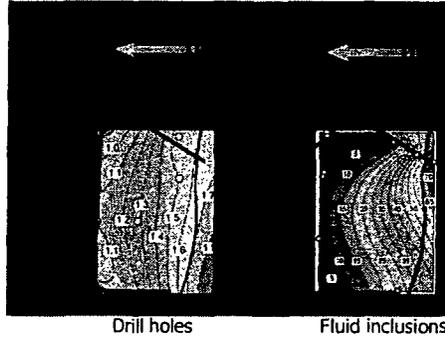


Calculated paleo  
heat flow

Yellow circles show  
locations of samples

**Assumptions:**  
Surface topography unchanged  
 $T_{\text{surface}} = 15^{\circ}\text{C}$   
 $k = 1.74 \text{ Wm}^{-1}\text{K}^{-1}$

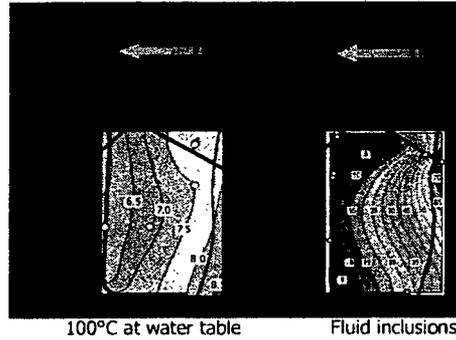
Gradients of the contemporary and paleo  
heat flows at Yucca Mountain



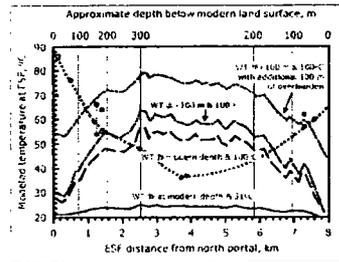
Nature of thermal gradients

Could these gradients be induced by the  
Timber Mountain Caldera hydrothermal  
episode ?

### Gradients during the Timber Mountain Caldera hydrothermal episode



### Model Timber Mountain Caldera temperatures vs. fluid inclusion temperatures



Fluid inclusion temperatures:

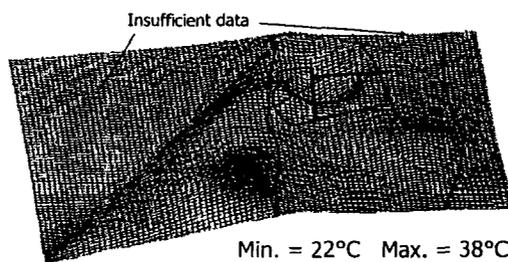
Blue – by Whelan et al. (2001)  
 Red – by Dublyansky et al. (2001)

Modified from Whelan et al. (2001)

### Paleo heat flow: Conclusions

- ❑ Values of paleo heat flow indicated by fluid inclusions are significantly greater than it is possible for net conductive heat transfer;
- ❑ Neither values nor spatial structure of paleo heat flow can be accounted for by any known event in thermal history of Yucca Mountain;
- ❑ Structure of paleo heat flow (steep east-west gradient) requires source of heat associated with the major block-bounding Paintbrush fault;

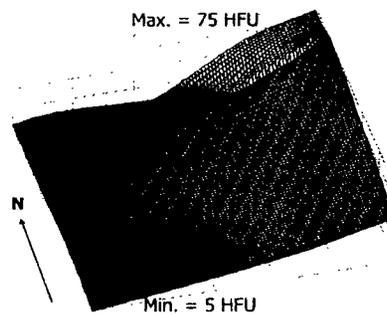
### Contemporary temperature at water table at Yucca Mountain



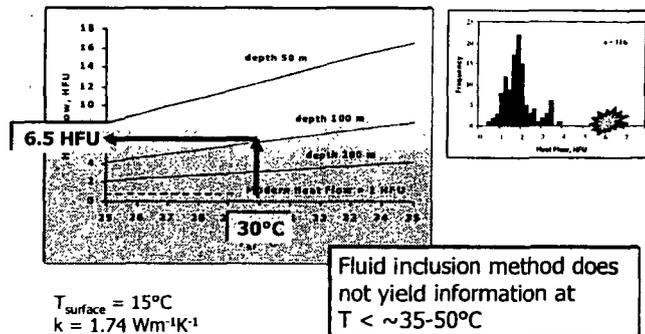
### Paleo heat flow: Conclusions (cont.)

- ❑ Parameters of the paleo heat flow preclude any substantial role of rainwater in the deposition of secondary minerals at Yucca Mountain;
- ❑ Extremely steep lateral heat gradient cannot be maintained for geologically significant periods of time, which again calls into the question the results of the radiometric age dating of secondary minerals at Yucca Mountain.
- ❑ This observation can only be explained by assuming short-lived transient character of heat input(s).

### Configuration of paleo heat flow at Yucca Mountain



### Significance of all-liquid inclusions: Do they indicate ambient temperature?



### Significance of all-liquid inclusions

"...FIAs with 2-phase fluid inclusions are not present in outermost Mg-enriched calcite that began precipitating in the LCZ between 3.8 and 1.9 Ma. Therefore, passage of fluids with elevated temperatures in this part of the repository site occurred prior to 1.9 Ma."

(Wilson et al., 2000)

"... results indicate that fluids with elevated temperatures were not present in the recent past (i.e.  $\leq 1.5$  Mya), but moved through the site more than 1.9 to  $\sim 2.8$  Ma and in some samples more than 4 to 5.3 Mya."

(Wilson and Cline, 2001)

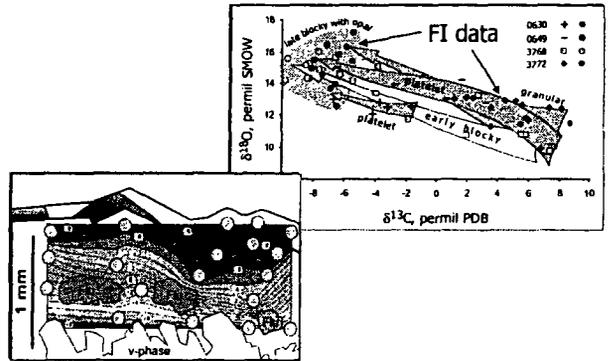
### Significance of all-liquid inclusions: Conclusions

- ❑ At a depth of the planned repository horizon, the temperature of  $\sim 35-50^{\circ}\text{C}$  may indicate either "ambient temperature" water or thermal water;
- ❑ Therefore, the conclusion regarding a non-thermal origin of the Mg-enriched calcite cannot be substantiated on the basis of the absence of the two-phase fluid inclusions.

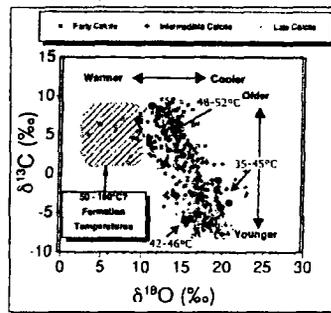
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### Monotonic evolution of isotopic properties



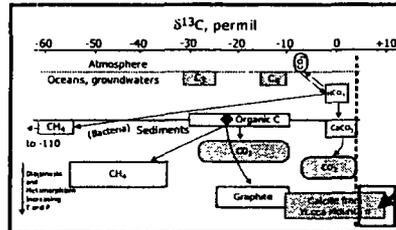
### Fluid inclusions and stable isotopes



Two-phase fluid inclusions are present in calcite with a range of isotopic properties, including those that are attributed to the youngest members of the paragenesis.

Modified from Whelan et al. (2001)

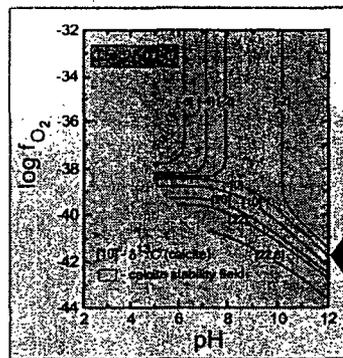
### Positive $\delta^{13}\text{C}$ in calcite



Positive (up to +9 per mil PDB)  $\delta^{13}\text{C}$  is incompatible with any known source of carbon at Yucca Mountain, and with the UZ zone environment, in general.

Principal sources of carbon in the near-surface environment (modified from Ohmoto, 1986).

### Positive $\delta^{13}\text{C}$ in calcite: Theory



Low fugacity of  $\text{O}_2$  allows preferential partitioning of  $^{12}\text{C}$  into the  $\text{CH}_4(\text{aq})$  that makes possible the deposition of calcite with positive  $\delta^{13}\text{C}$

$\delta^{13}\text{C}$  contours in the stability field of calcite @  $I = 1$  and  $\delta^{13}\text{C}_{\text{carbon}} = -5$  ‰ PDB (Ohmoto, 1972)

#### Overall conclusion

- ❖ Observations presented above cannot reasonably be explained by a model invoking deposition of secondary minerals at Yucca Mountain from percolating rainwater;
- ❖ A model, which explains all observations presently known to us is the model of upwelling.

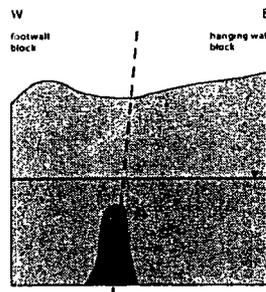
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### Requirements for a conceptual model

- ❖ Consistent with basic sciences, such as physics and chemistry;
- ❖ Contradiction-free;
- ❖ Coherently explains all observations.

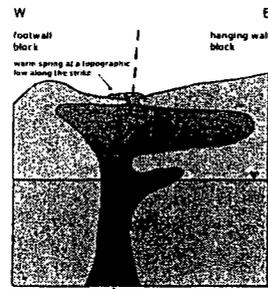
### Development of the hydro-tectonic disturbance: a model



#### ❖ Stage 1:

Upward flow initiated by an earthquake ( $M \sim 7$ ) with a hypocenter at a depth of 10-15 km on a deep seated fault

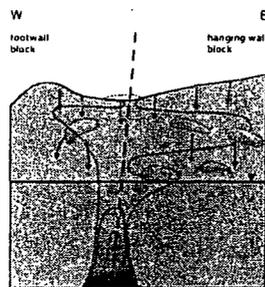
### Development of the hydro-tectonic disturbance: a model



#### Stage 2:

Formation of a transient thermal water mound within the vadose zone; lateral flow in the enhanced permeability zones

### Development of the hydro-tectonic disturbance: a model



#### Stage 3

Decay of the mound; formation of perched water bodies; downward flow; interaction with rainwater.